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(54) Title: AMMONIA OXIDATION CATALYSTS			
(57) Abstract			
<p>An ammonia oxidation catalyst <u>cartridge</u> unit comprises a pair of retaining meshes spaced apart by less than 50 mm defining an enclosure filled with a random packed mass of particles of a composition comprising oxides of at least one metal selected from manganese, iron, nickel and cobalt, especially cobalt and at least one element A selected from rare earths and yttrium in such proportions that the element A to cobalt atomic ratio is in the range of 0.8 to 1.2. The catalyst particles have a minimum dimension of at least 0.5 mm and a maximum dimension of not more than 5 mm, and the spacing between the meshes is at least 4 times, but not more than 50 times, the maximum dimension of the particles. The unit may extend across the ammonia oxidation reactor cross section or a catalyst cartridge may comprise a grid or apertured plate, extending across the cross section of the reactor, with a unit as aforesaid in each aperture.</p>			

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Ammonia oxidation catalysts

This invention relates to ammonia oxidation catalysts. Ammonia oxidation is widely employed in the manufacture of nitric acid and hydrogen cyanide. In the manufacture of nitric acid ammonia is oxidised with air to nitric oxide, while in the manufacture of hydrogen cyanide a mixture 5 of ammonia and methane (often as natural gas) is oxidised with air. In both processes, the gas mixture is passed at an elevated temperature over a catalyst to effect the oxidation. Side reactions, such as the formation of nitrogen or nitrous oxide, are undesirable. Consequently, in addition to good activity, the catalyst is required to have a good selectivity.

For many years the catalysts employed have been platinum, sometimes alloyed with other 10 precious metals, in the form of meshes or gauzes formed from the metal wire. Such catalysts have good activity and selectivity but suffer from the disadvantage that not only is the catalyst very expensive, but at the temperatures encountered, the metals exhibit an appreciable volatility and so gradually the metal is lost into the gas stream. While it is well known to provide downstream means to trap the volatilised metal so that it may be recovered subsequently, because of the continual 15 volatilisation, the life of the catalyst is short and frequent replacement is necessary. Furthermore the recovery of the metals from the downstream trap and re-fabrication of the catalyst meshes or gauzes involves a considerable allocation of working capital.

It is therefore desirable to provide a replacement for such precious metal catalysts.

It is well known that oxides of metals such as manganese, iron, nickel or, especially cobalt, 20 often in conjunction with a one or more rare earth oxides, exhibit activity for ammonia oxidation. Thus US 3 888 792 discloses ammonia oxidation using catalyst compositions comprising cobalt oxide and a small proportion of a rare earth oxide. US 4 812 300 discloses ammonia oxidation using catalysts such as LaCoO_3 said to have the Perovskite structure. CN-A-86 108 985 discloses ammonia oxidation using lanthana/ceria/cobalt oxide compositions of the general formula 25 $\text{La}_{1-x}\text{Ce}_x\text{CoO}_3$ (where x is from 0 to 1) made by a specified co-precipitation route, and indicates that such materials had good activity and selectivity when tested on a small scale, although there is some suggestion that the activity and/or selectivity is decreased at operating temperatures in the upper end of the temperature range (800-1000°C) normally employed for ammonia oxidation.

Ammonia oxidation is normally operated by passing the heated ammonia/air mixture at a 30 high linear velocity (measured at NTP) through a wad of precious metal meshes or gauzes extending across the cross-section of the reactor. Commercial ammonia oxidation plants usually employ reactors having a diameter in the range 0.5 to 5 m. Linear gas velocities conventionally employed in commercial ammonia oxidation processes operating substantially at atmospheric pressure are usually in an excess of 2000 m/h, and typically are in the range 5000-10000 m/h, 35 preferably 4000-6000 m/h. In plants operating at higher pressures, e.g. up to 10 barg., the linear velocities are generally in the range 20000 to 50000 m/h.

Desirably the catalyst should be in a form such that it is a direct replacement for the precious metal meshes or gauzes conventionally used. Thus it is desirable to provide the catalyst bed in the form of a cartridge that can replace the conventional wad of meshes or gauzes.

Another factor of importance is the temperature to which the feed gas must be pre-heated in order that the oxidation process may sustain itself. For a given set of processing conditions, i.e. catalyst particle size, catalyst bed depth, gas composition and linear flow rate, if the inlet temperature is decreased, at a certain value the oxidation reaction no longer can sustain itself. This temperature is termed the "blow out" temperature. For normal commercial operation it is desirable that the "blow-out" temperature is below 250°C, particularly below 150°C. We have found that for a given gas composition and gas linear flow rate, the "blow-out" temperature decreases as the particle size of the catalyst decreases and as the depth of the catalyst bed increases. Hence from the "blow-out" temperature considerations, the bed depth should be as great as possible and the particle size as small as possible. Unfortunately, yet another factor of importance is the pressure drop experienced by the gas as it passes through the catalyst bed. This increases as the particle size of the catalyst is decreased and as the catalyst bed depth increases, and so a compromise is necessary to achieve an acceptable "blow-out" temperature while at the same time have an acceptable pressure drop. We have found that for catalysts of a convenient particle size, the bed depth should be less than 50 mm. At a linear gas velocity of 4000 m/h through a catalyst bed of depth 40 mm, the space velocity is 10^5 h^{-1} . We have found that space velocities above 10^5 h^{-1} are normally required.

In the aforesaid US 3 888 792 the catalysts in the form of a random packed mass of cylindrical pellets, or extrudates of 4 mm diameter and 4 mm length, were tested using bed depths ranging from 20 to 60 mm. In the aforesaid CN-A-86 108 985 the catalysts in the form of a coarse powder were tested using a bed depth of 24 mm and, in the form of cylindrical tablets of 6.5 mm diameter and 4 mm length, using bed depths ranging from 40 to 80 mm at space velocities of the order of 3×10^4 to $1 \times 10^5 \text{ h}^{-1}$.

As indicated above it is desirable to provide the catalyst bed in the form of a cartridge that can replace the conventional wad of meshes or gauzes. The provision of a catalyst cartridge of diameter suitable for use in a conventional ammonia oxidation reactor, and having a catalyst bed depth of less than 50 mm, presents problems of maintaining the catalyst bed depth uniform.

In the present invention this is overcome by disposing the catalyst particles between a pair of retaining meshes.

Accordingly the present invention provides an ammonia oxidation catalyst bed cartridge unit comprising a pair of retaining meshes spaced apart by less than 50 mm defining an enclosure filled with a random packed mass of particles of a composition comprising oxides of at least one metal selected from manganese, iron, nickel and cobalt, said particles having a minimum dimension of at

least 0.5 mm and a maximum dimension of not more than 5 mm, the spacing between said meshes being at least 4 times, and less than 50 times, the maximum dimension of the particles.

In some cases it may be desirable to employ a catalyst bed having different size particles: for example it may be desirable that the particles in the inlet region of the bed are larger than those 5 in the outlet region. In such a case the cartridge may comprise three spaced gauzes with the smaller particles between the middle, or intermediate, gauze and the lower gauze and the larger particles between the upper and intermediate gauzes. Where the bed is formed of different size particles as aforesaid with an intermediate gauze, the intermediate gauze is disregarded in determining the spacing of the gauzes.

10 It will be appreciated that the meshes, which may be of a suitable heat resistant metal such as stainless steel, should have as large an aperture as is consistent with retaining the particles on the mesh. The meshes are preferably catalytically inert.

The cartridge unit may comprise a pair of spaced apart mesh sheets dimensioned so as to fit across the reactor. The mesh sheets may be held spaced apart by a suitable spacing member 15 disposed round the periphery of the unit. Spacing members may also be disposed at intervals across the unit. The unit is preferably dimensioned such that it is a snug fit inside the ammonia oxidation reactor and extends across the cross-section of the reactor. Alternatively the cartridge may comprise a supporting unit, for example a grid or apertured plate dimensioned so as to fit across the reactor with a cartridge sub-unit positioned in each aperture of the plate or grid, with 20 each sub-unit comprising a pair of spaced apart meshes with the catalyst disposed between the meshes.

The catalyst particles may be irregular granules or may be moulded or formed into geometric shapes, such as cylinders. Preferred catalyst particles are moulded cylindrical tablets having a diameter in the range 0.5 to 4 mm and a length of 0.5 to 4 mm. The aspect ratio of the particles, i.e. 25 the ratio of the maximum dimension to the minimum dimension, is preferably less than 2. Moulded particles, e.g. cylinders, may have one or more passages therethrough to increase the geometric surface area and to increase the bed voidage thereby reducing the pressure drop across the bed. The bed depth, i.e. the spacing between the meshes, is preferably 5 to 20 times the maximum dimension of the shaped particles.

30 By the use of particles of the specified size and shape, e.g. the provision of through passages, a high geometric surface area of catalyst is exposed to the reactants but, by having the specified spacing between the meshes, bypassing of the catalyst is minimised while maintaining a sufficiently thin bed that an acceptable pressure drop will be achieved at high space velocities.

The catalyst preferably contains at least one mixed oxide phase containing cobalt and at 35 least one element A selected from rare earths and yttrium. The element A to cobalt atomic ratio is preferably 0.8 to 1.2, particularly 1.0 to 1.2. Preferably at least one element selected from yttrium, cerium, lanthanum, neodymium, and praseodymium is used as part or all of element A. Element A

may comprise a mixture of at least one variable valency element Vv selected from cerium and praseodymium and at least one non-variable valency element Vn selected from yttrium and the non-variable valency rare earth elements such as lanthanum or neodymium. In particular it is preferred that the atomic proportions of variable valency element Vv to non-variable valency element Vn is in the range 0 to 1, particularly 0 to 0.3.

5 As indicated above, the catalyst may be in a form wherein the amount of oxygen is non-stoichiometric. This arises from the variable valency of cobalt and also of any variable valency rare earth present as part, or all, of element A.

The catalyst particles may be made by forming a finely divided powder composition into
10 particles of the desired size by techniques such as granulation, extrusion or moulding. The finely divided powder composition may be made by precipitation by adding a solution of soluble salts of the relevant metals to a solution of a base, e.g. ammonium carbonate or hydroxide, to precipitate the relevant metals as (basic) carbonates, hydroxides, or oxides followed by calcination to convert the precipitated compounds to the oxides. The use of alkali metal compounds as the base to effect
15 precipitation is less preferred as they inevitably cause some contamination of the product with sodium which could act as a catalyst poison. The precipitation may alternatively, but less preferably, be effected by adding the base to the solution of the mixed salts. Where the active metal is cobalt, it is preferred to employ a continuous precipitation method at a controlled pH. This is because cobalt forms soluble ammine compounds at high pH in aqueous ammoniacal solutions
20 and so precipitation at a controlled pH below that at which such ammines are formed is desirable to avoid loss of cobalt. Alternatively, the finely divided powder composition may be made by forming a solution of thermally decomposable salts, e.g. nitrates or salts of organic acids, e.g. oxalates or citrates, of the metals in the appropriate proportions and evaporating the solution to dryness followed by calcination to effect decomposition to the appropriate oxides. Less preferably, the
25 composition may be made by mixing preformed oxides of the metals in the appropriate proportions.

In another alternative, the active material, e.g. cobalt may be present as a coating on suitable support particles. Thus a finely divided oxidic material, e.g. ceria, may be impregnated with a solution containing a cobalt salt, and possibly also salts of other elements such as a salt of an element A, e.g. a lanthanum salt, followed by decomposition of the salts. Alternatively, such a
30 supported material may be made by precipitation by precipitating the cobalt, and optionally other elements, as heat decomposable compounds on to a finely divided, e.g. precipitated, oxidic support compound or compound decomposable thereto.

Whichever route is used to make the finely divided powder composition, the composition should be calcined, e.g. in air, preferably at a temperature in the range 900-1200°C, before or
35 preferably after granulating, extruding or moulding into particles of the desired size and shape.

The invention is illustrated by the accompanying drawings (which are not to scale) in which:

Figure 1 is a diagrammatic cross section of a unit in accordance with a first embodiment of the invention

Figure 2 is a diagrammatic cross section of a part of unit in accordance with a second embodiment of the invention

5 Figure 3 is a diagrammatic cross section through an ammonia oxidation reactor incorporating a unit of the present invention

Figure 4 is a plan view of part of a wire gauze showing cuts and fold lines

Figure 5 is a sketch of part of a gauze as shown in Figure 4 folded to form a compartmentalised structure.

10 Figure 6 is a diagrammatic cross section of a unit in accordance with a third embodiment of the invention

Figure 7 is an enlargement of part of Figure 6 showing the configuration of the cartridge sub-units of Figure 6.

Figure 8 is a cross section of a reactor showing the support grid and cartridge configuration of a fourth embodiment of the invention

15 Figure 9 is a sketch of one of the cartridge units of Figure 8

Figure 10 is a section through the cartridge unit of Figure 9.

In Figure 1 there is shown a catalyst cartridge consisting of a pair of circular stainless steel meshes 1, 2 of diameter about 3 m having a mesh size of for example about 2 mm held spaced 20 mm apart by a spacer ring 3. Additional spacer members 4 are disposed at intervals across the cartridge. The space enclosed by the meshes and the ring is filled with random packed cylindrical catalyst tablets of 3 mm diameter and 3 mm height.

When using beds of the type shown in Figure 1, there is a risk that the gauzes, particularly the lower gauze, may stretch and/or sag after exposure to the prevailing operating conditions for an 25 extended period of time. As a result the catalyst particles may re-distribute themselves so that some areas of the bed have a lesser bed depth than others. This is undesirable as the process gas will preferentially pass through the portions of lesser bed depth and insufficient reaction may occur. It is therefore preferred that the lower gauze is supported by a rigid porous structure such as tiles of a ceramic foam material.

30 To minimise the re-distribution of the catalyst particles, the upper mesh may be held firmly against the bed of catalyst particles. Thus, as shown in the embodiment of Figure 2, rivets 5 may be provided extending through the meshes 1, 2 and the catalyst bed, and a spring 6 provided above the upper mesh 1 engaging with the upper mesh 1 and a cap 7 on the upper end of the rivet. The spring is preferably located on the upstream side of the catalyst bed, since the temperature in 35 operation is lower than that on the downstream side of the catalyst bed. The rivets 5 may be provided in addition to, or instead of, the intermediate spacers 4 of the embodiment of Figure 1.

In Figure 3 there is shown an ammonia oxidation reactor 10 having an inlet port 11 and an outlet port 12. Disposed across the reactor is a catalyst cartridge 13, of the type shown in Figure 1 supported by ceramic foam tiles 14 resting on cross members 15 extending across the reactor 10.

Alternatively, or additionally, re-distribution of the catalyst particles within the bed may be minimised by dividing the bed into small compartments. Thus the lower gauze may be cut and folded to form upstanding dividing walls, preferably of height substantially equal to the desired bed depth. Thus as shown in Figures 4 and 5 a sheet of wire gauze 16 is cut along the lines 17, 18, 19 and 20 and then folded in alternate directions along the dotted lines a-a, b-b and c-c, and along lines d-d, e-e and f-f and d'-d', e'-e' and f'-f' etc. to form upstanding walls 21, 22, 23 and 24. The flaps e.g. 25 formed by the cuts 17 to 20 are then tack welded to the upstanding walls. The compartments are then filled with the particulate catalyst. The compartments may be of any convenient shape, e.g. square, rectangular, triangular etc.

As indicated above in some cases it may be desirable to employ a catalyst bed having different size particles by providing the cartridge with three spaced gauzes with the smaller particles between the middle gauze and the lower gauze and the larger particles between the upper and middle gauzes. In such a case the lower gauze may be shaped to give compartments as shown in Figures 4 and 5 while the upper portion of the bed, i.e. containing the larger particles, is a continuous bed.

In Figures 6 and 7 there is shown an alternative construction for the cartridge. This construction is particularly suited to embodiments where the catalyst particle size is relatively small, typically maximum dimension below 2 mm, and the bed depth shallow, e.g. less than 25 mm. The cartridge consists of a circular plate 26 dimensioned to fit across the reactor, e.g. in place of cartridge 13 of Figure 3. The plate has a plurality of circular apertures, e.g. of 100 mm diameter, spaced apart at e.g. 200 mm centres. Located within each aperture is a cartridge sub-unit 27. Each sub-unit consists of a plate 28 having a central aperture 29 and a pair of concentric cylindrical meshes 30, 31, typically of length 200-300 mm, fastened at one end to the plate 28. The other end of the meshes is fastened to a closure plate 32. The spacing between the concentric meshes is typically 10-20 mm and the catalyst particles are disposed in the space between the two meshes 30, 31.

By means of the construction of the second embodiment, it is possible, by suitably dimensioning the size and spacing of the sub-units 27 and the length of the concentric meshes 30, 31, to provide a gas flow area through the catalyst that is significantly greater than the cross-sectional area of the reactor. This means that although the linear velocity of gas through the reactor may be similar to that employed with the cartridge of the type shown in Figure 1, the depth of the catalyst bed can be decreased, and hence the pressure drop reduced, without increasing the space velocity.

In yet another embodiment as shown in Figures 8 to 10, a square grid of cartridge supports 33 is disposed across the reactor and a catalyst cartridge 34 is disposed in each square of the grid. It will be appreciated that the grid, and hence the cartridges need not be square, but may be of another polygonal shape, e.g. rectangular or triangular, in plan. The spaces 35 in the grid at 5 the edges of the reactor that are not large enough to receive a cartridge 34 are closed with blanking members. Alternatively cartridges of a non-standard size may be fitted in these spaces. Each cartridge 34 is in the form of an inverted pyramid structure having a flange 36 by which the cartridge may be supported on the grid cartridge supports 33. The pyramid consists of an outer hollow square base 37 from which angle members 38 defining the outer inclined edges of the pyramid are 10 disposed. Triangular ceramic foam tiles 39, forming the inclined faces of the pyramid, are supported by the angle members 38. These foam tiles 39 serve to support a lower, inverted pyramidal-shaped, mesh or gauze 40 attached at its upper, base, end to the outer square base 37. Supported within the outer hollow square base 37 by means of support brackets 41 spaced at intervals around the outer base 37 is a hollow inner square base 42 from which depends a second, 15 inner, inverted pyramidal-shaped, mesh or gauze 43. A spacer member 44 is disposed at the apex of the pyramid to hold the two gauzes apart. The catalyst particles are disposed in the space 45 between the two gauzes. The space 46 between the walls of the outer and inner square bases 37 and 42 provides a reservoir for catalyst particles so that the space 45 between the gauzes 40, 43 may be kept full of catalyst particles in the event of settlement of the catalyst particles during use.

20 It is seen that in use, the process gas flows down through the inner hollow base 42, through inner gauze 43, through the catalyst-filled space 45, and then through outer gauze 40 and the ceramic foam tile 39 as shown by the arrows.

This form of cartridge construction facilitates replacement of the cartridges in the reactor and charging and discharging catalyst from the individual cartridges. It also has the benefit that 25 optimum use may be made of the reactor cross sectional area while providing for a catalyst bed area substantially greater than that of the reactor cross sectional area.

The catalyst units or cartridges of the invention may be used as a direct replacement for the conventional precious metal catalysts with essentially no modification to the ammonia oxidation process, except of course the conventional precious metal trap arrangements can be eliminated. In 30 the oxidation of ammonia to nitric oxide for the manufacture of nitric acid, the oxidation process may be operated at temperatures of 800-1000°C, particularly 850-950°C, pressures of 1 to 15 bar abs., with ammonia in air concentrations of 5-15%, often about 10%, by volume.

In addition for use for ammonia oxidation reactions, the catalyst units may also be of use for other oxidations.

35 The invention is illustrated by the following examples.

Example 1

A finely divided lanthana/ceria/cobalt oxide catalyst (La:Ce:Co atomic proportions 8:2:10) was made by precipitation followed by calcination at 900°C. The resultant powder was formed into cylindrical pellets of 3 mm diameter and 3 mm height. A preheated mixture of air and ammonia containing about 10% by volume of ammonia was passed at atmospheric pressure through a bed of 40 mm depth of the pellets at various linear velocities. The preheat temperature was gradually decreased and the temperature ("blow-out" temperature) noted at which the reaction no longer was self-sustaining. While the reaction was self-sustaining the temperature of the gas mixture leaving the catalyst bed was typically about 600-650°C higher than the preheat temperature. The procedure was repeated with a catalyst bed of 20 mm depth. The results are shown in the following table.

Linear gas velocity (m/h)	"Blow-out" temperature (°C)	
	Bed depth 20 mm	Bed depth 40 mm
2350	97	-
2900	-	76
3500	117	90
4700	148	115
5900	185	130
7100	197	165

Example 2

The procedure of Example 1 was repeated but with the catalyst in the form of granules of diameter 0.5 mm and 2 mm, and cylindrical pellets as used in Example 1. In this example beds of various depths were used at a constant linear gas flow rate of 4500 m/h. The results are shown in the following table.

Bed depth (mm)	"Blow-out" temperature (°C)		
	0.5 mm granules	2 mm granules	3 mm pellets
2	95	-	-
4	80	155	-
6	55	-	-
9	45	-	-
10	-	135	-
20	-	-	150
30	-	-	110
40	-	-	100

Claims

1.

An ammonia oxidation catalyst bed cartridge unit comprising a pair of retaining meshes spaced apart by less than 50 mm defining an enclosure filled with a random packed mass of particles of a composition comprising oxides of at least one metal selected from manganese, iron, nickel and cobalt, said particles having a minimum dimension of at least 0.5 mm and a maximum dimension of not more than 5 mm, the spacing between said meshes being at least 4 times, and less than 50 times, the maximum dimension of the particles.

2.

A unit according to claim 1 wherein the catalyst particles are moulded cylindrical tablets having a diameter in the range 0.5 to 4 mm and a length of 0.5 to 4 mm.

3.

A unit according to claim 1 or claim 2 wherein the catalyst particles have an aspect ratio of less than 2.

4.

A unit according to any one of claims 1 to 3 wherein the spacing between the meshes is 5 to 20 times the maximum dimension of the particles.

5.

A unit according to any one of claims 1 to 4 wherein the catalyst particles comprise oxides of (a) at least one element A selected from rare earths and yttrium, and (b) cobalt, said cobalt and element A being in such proportions that the element A to cobalt atomic ratio is in the range 0.8 to 1.2.

6.

A unit according to claim 5 wherein part or all of element A is at least one element selected from yttrium, cerium, lanthanum, neodymium, and praseodymium.

7.

A unit according to claim 6 wherein element A comprises a mixture of at least one variable valency element Vv selected from cerium and praseodymium and at least one non-variable valency element Vn selected from yttrium and a non-variable valency rare earth element.

8.

A unit according to claim 7 wherein the atomic proportions of variable valency element Vv to non-variable valency element Vn is in the range 0 to 1.

9.

A unit according to any one of claims 1 to 8 wherein the space between the meshes is divided into compartments by means of integral walls formed by folding one of the meshes.

10. A unit according to any one of claims 1 to 9 wherein the meshes are each of inverted pyramidal-configuration disposed one within the other with the catalyst particles disposed in the space between the two meshes.
11. A cartridge comprising a plate having a plurality of apertures therein with a unit according to 10 disposed in each aperture.
12. A cartridge comprising a plate having a plurality of apertures therein with a unit according to any one of claims 1 to 9 disposed in each aperture, each unit having the retaining meshes as a pair of concentric cylinders with the catalyst particles disposed in the annular space between the concentric meshes.
13. An ammonia oxidation process comprising feeding ammonia and air to a reactor and passing the resultant air/ammonia mixture, at an elevated temperature and at a space velocity (at NTP) of at least $1.2 \times 10^5 \text{ h}^{-1}$ through a cartridge unit according to any one of claims 1 to 10, or a cartridge according to claim 11 or claim 12, extending across the cross section of the reactor.

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Fig.1.

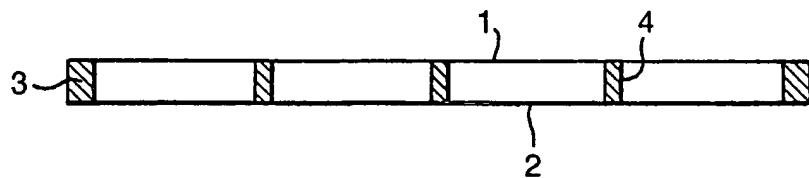


Fig.2.

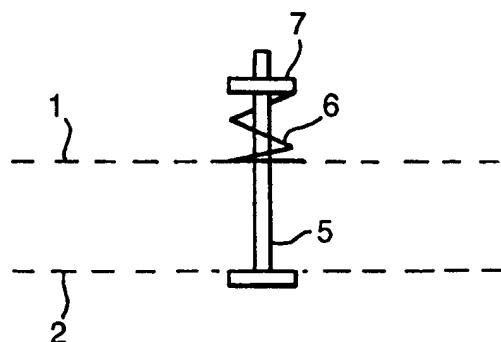


Fig.3.

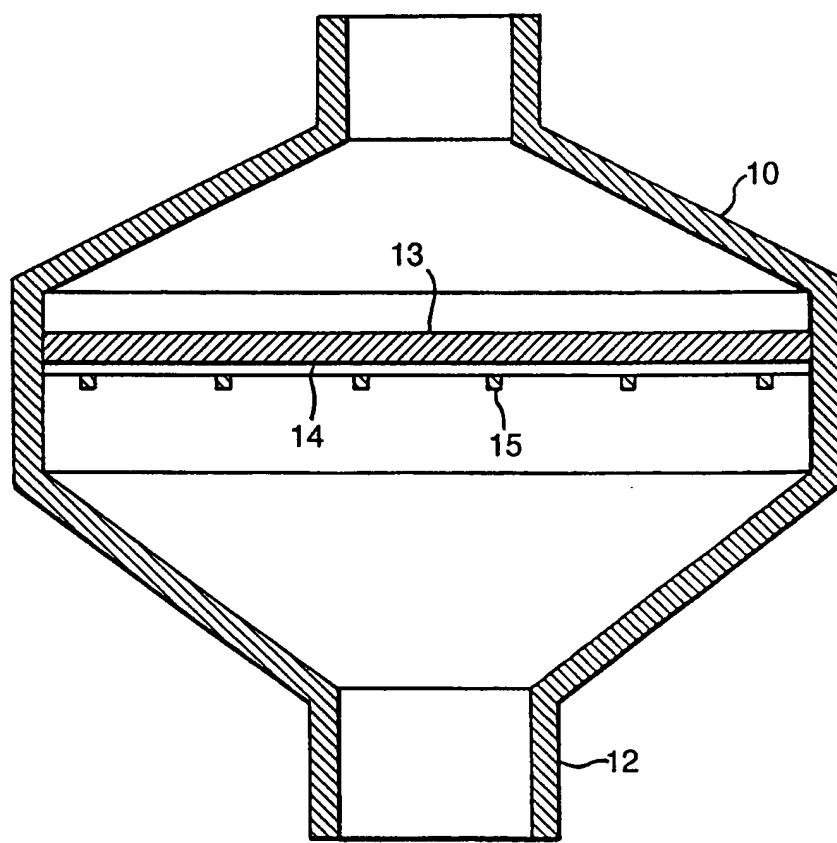


Fig.4.

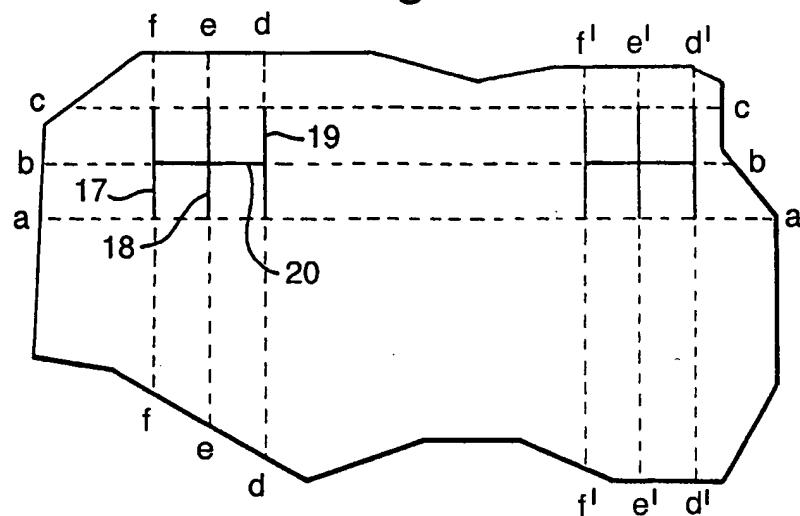
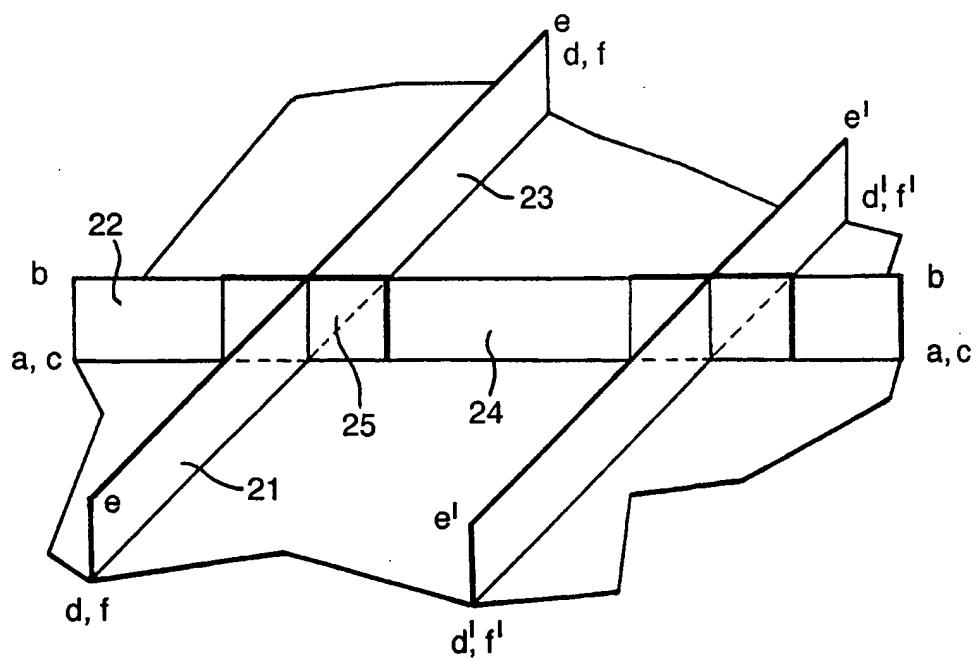


Fig.5.



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Fig.6.

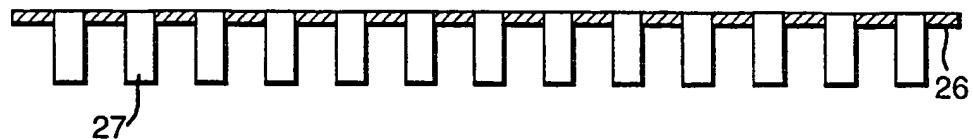


Fig.7.

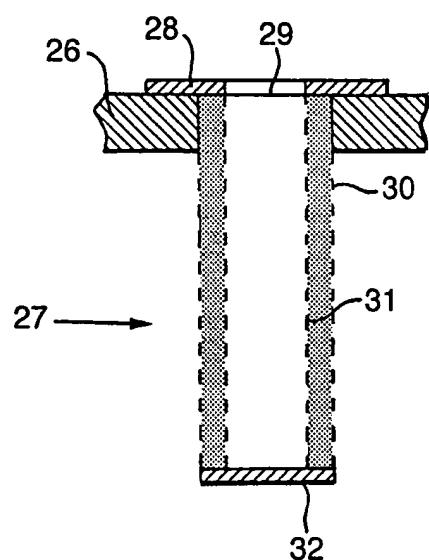
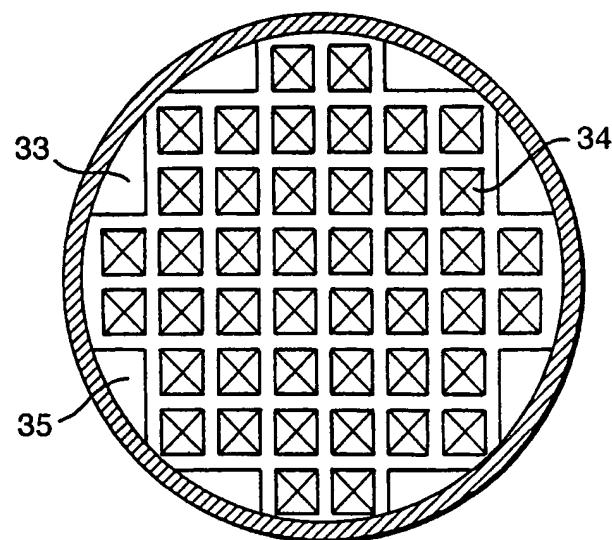


Fig.8.



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Fig.9.

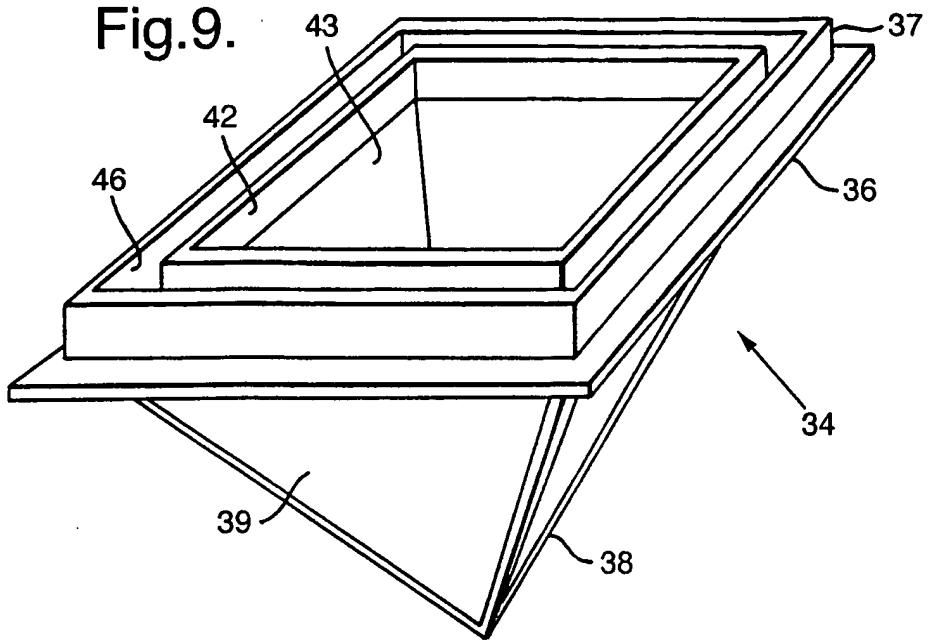
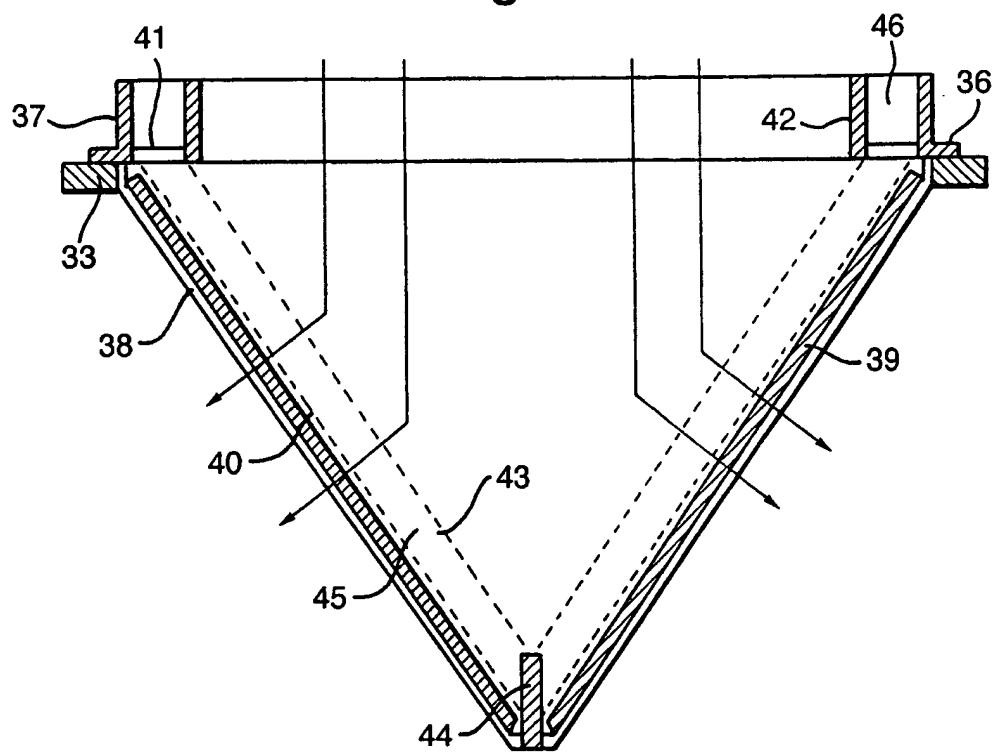


Fig.10.



INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 98/03386

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 6 C01B21/26 C01B21/28 B01J8/02 B01J23/83

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
 IPC 6 C01B B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE 21 48 707 A (ICI) 22 June 1972 see the whole document	1-7, 13
Y		7-9, 12, 13
Y	----- CHEMICAL ABSTRACTS, vol. 111, no. 22, 27 November 1989 Columbus, Ohio, US; abstract no. 202883, WU, YUE ET AL: "Lanthanum cerium cobalt oxide catalyst for ammonia oxidation" XP002092533 see abstract & CN 86 108 985 A (CHINESE ACADEMY OF SCIENCES, CHANGCHUN INSTITUTE OF APPLIED CHEMISTRY,) ----- -/-	7, 8

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
8 February 1999	18/02/1999
Name and mailing address of the ISA	Authorized officer

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PCT/GB 98/03386

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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